

Emergence of Fully-Gapped s_{++} -wave and Nodal d -wave States Mediated by Orbital- and Spin-Fluctuations in Ten-Orbital Model for KFe_2Se_2

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We study the superconducting state in recently discovered high- T_c superconductor $\text{K}_x\text{Fe}_2\text{Se}_2$ based on the ten-orbital Hubbard-Holstein model without hole-pockets. When the Coulomb interaction is large, spin-fluctuation mediated d -wave state appears due to the nesting between electron-pockets. Interestingly, the symmetry of the body-centered tetragonal structure in $\text{K}_x\text{Fe}_2\text{Se}_2$ requires the existence of nodes in the d -wave gap, although fully-gapped d -wave state is realized in the case of simple tetragonal structure. In the presence of moderate electron-phonon interaction due to Fe-ion optical modes, on the other hand, orbital fluctuations give rise to the fully-gapped s_{++} -wave state without sign reversal. Therefore, both superconducting states are distinguishable by careful measurements of the gap structure or the impurity effect on T_c .

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The pairing mechanism of high- T_c iron-based superconductors has been significant open problem. The main characters of FeAs compounds would be (i) the nesting between electron-pockets (e-pockets) and hole-pockets (h-pockets), and (ii) the existence of orbital degree of freedom. By focusing on the intra-orbital nesting, fully-gapped sign-reversing s -wave state (s_{\pm} -wave state) had been predicted based on the spin fluctuation theories^{1,2}. On the other hand, existence of moderate electron-phonon (e -ph) interactions due to Fe-ion optical phonons and the inter-orbital nesting can produce large orbital fluctuations³. Then, orbital-fluctuation-mediated s -wave state without sign reversal (s_{++} -wave state) had been predicted by using the random-phase-approximation (RPA)^{3,4} or the fluctuation-exchange (FLEX) approximation⁵. According to the analysis in Refs. 6,7 the s_{++} -wave state is consistent with the robustness of T_c against randomness^{8,9} as well as the “resonance-like” hump structure in the neutron inelastic scattering¹⁰. Non-Fermi liquid transport phenomena in ρ ¹¹ can be explained by the development of orbital fluctuations⁵.

Recently, iron-selenium 122-structure compound $\text{A}_x\text{Fe}_2\text{Se}_2$ (A = alkaline metals) with $T_c \sim 30$ K was discovered¹². This heavily electron-doped superconductor has been attracting great attention since both the band calculations^{13,14} and angle-resolved-photoemission-spectrum (ARPES) measurements^{15–17} indicate the absence of h-pockets. NMR measurements reports the weakness of spin fluctuations¹⁸, and both ARPES^{15–17} and specific heat measurements¹⁹ indicate the isotropic SC gap. Thus, study of $\text{A}_x\text{Fe}_2\text{Se}_2$ will give us important information to reveal the pairing mechanism of iron pnictides.

The unit-cell of iron-based superconductors contains two Fe atoms. However, except for 122-systems, one can construct a simple “single-Fe model” from the original “two-Fe model” by applying the gauge transformation on d -orbitals²⁰. By this procedure, the original

Brillouin zone (BZ) is enlarged to the “unfolded BZ”. Based on the single-Fe model, spin-fluctuation-mediated d -wave state (B_{1g} representation) “without nodes” had been proposed^{21–23}, by focusing on the nesting between e-pockets. However, we cannot construct a “single-Fe model” for 122 systems since finite hybridization between e-pockets prevents the unfolding procedure²⁰. Therefore, theoretical study based on the original two-Fe model is highly desired to conclude the gap structure.

In this paper, we study the ten-orbital (two Fe atoms) Hubbard-Holstein (HH) model for KFe_2Se_2 using the RPA. When the Coulomb interaction is large, we obtain the d -wave SC state due to the spin fluctuations, as predicted by the recent theoretical studies in the single-Fe Hubbard models^{21–23}. However, the gap function on the Fermi surfaces (FSs) inevitably has “nodal structure” in the two-Fe model, due to the symmetry requirement of the body-centered tetragonal lattice. On the other hand, orbital-fluctuation-mediated s_{++} -wave state is realized by small e -ph coupling; the dimensionless coupling constant $\lambda = gN(0)$ is just ~ 0.2 . Since the nodal SC state is fragile against randomness, study of impurity effect will be useful to distinguish these SC states.

We perform the local-density-approximation (LDA) band calculation for KFe_2Se_2 using Wien2k code based on the experimental crystal structure¹². Next, we derive the ten-orbital tight-binding model that reproduces the LDA band structure and its orbital character using Wannier90 code and Wien2Wannier interface²⁴. The dispersion of the model and the primitive BZ are shown in Figs. 1 (a) and (b). Based on a similar ten-orbital model, Suzuki *et al.* studied the s_{\pm} -wave gap structure for BaFe_2As_2 ²⁵.

In Fig. 1, we show the FSs of KFe_2Se_2 for (c) $k_z = 0$ and (d) $k_z = \pi$ planes when the electron number per Fe-ion is $n = 6.5$: On each plane, there are four large and heavy e-pockets around X and Y points, and one small and light e-pockets around Z point. For $n = 6.5$, the energy of the h-band at Γ point from the Fermi level,

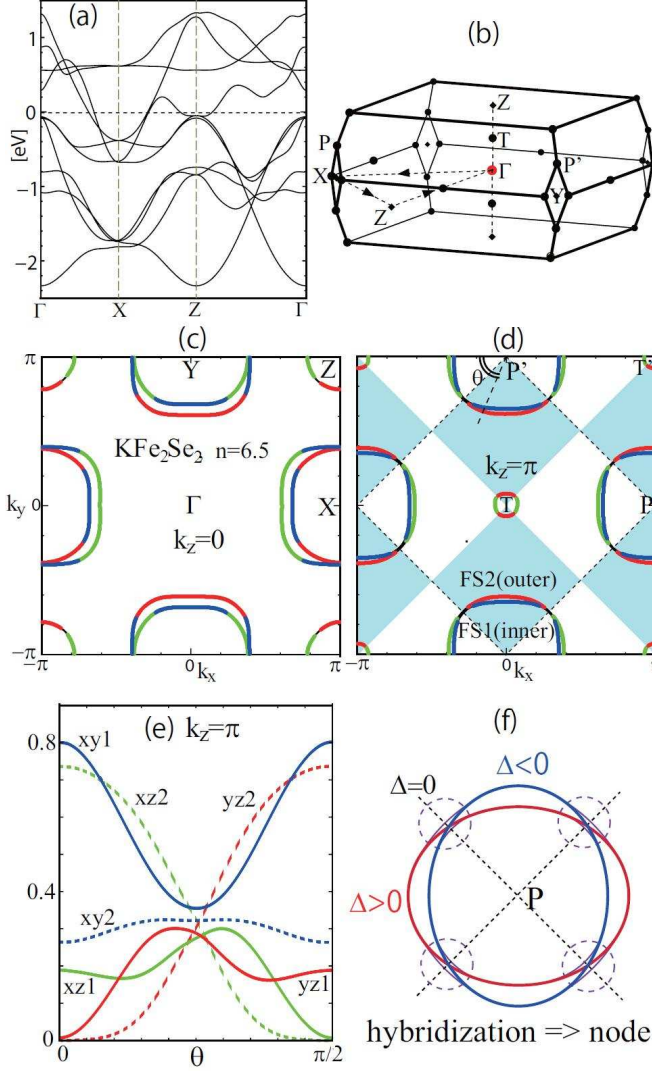


FIG. 1: (color online) (a) Dispersion of the present ten-orbital model for KFe_2Se_2 . Γ , X , and Z points are on the $k_z = 0$ plane. (b) Primitive BZ for body-centered tetragonal lattice. (c)(d) FSs on the $k_z = 0$ plane and $k_z = \pi$ plane. The green (light gray), red (gray), and blue (dark gray) lines correspond to xz , yz , and xy orbitals, respectively. The diamond-shaped shadows in (d) indicates the sign of basis function for B_{1g} representation. (e) Weight of each orbital on the inter FS (FS1) and the outer FS (FS2) as function of θ ; θ is shown in (d). (f) Hybridization between two e-pockets in 122 systems should create the nodal d -wave gap.

E_h , is about -0.07 eV. Since the obtained FS topology and the value of E_h are consistent with recent reports by ARPES measurements^{15–17}, we study the case $n = 6.5$ hereafter. In the present BZ in (b), Γ and Z points and X and Y points in (c) are not equivalent, and $k_z = 2\pi$ plane is given by shifting (c) by (π, π) . As for (d), T and T' points and P and P' points are equivalent, meaning that the reciprocal wave vector on the $k_z = \pi$ plane is (π, π) and $(\pi, -\pi)$. The diamond-shaped shadows in the $k_z = \pi$ plane indicates the sign of basis function for B_{1g}

($x^2 - y^2$ -type) representation, which has nodes on the P - P' line on both FS1 (inner FS) and FS2 (outer FS).

To confirm the existence of nodes, we verify that FS1 and FS2 in KFe_2Se_2 are largely hybridized. In fact, the weights of d -orbitals on FS1,2 given in Fig. 1 (e) are smooth functions of θ , which is the strong evidence for the hybridization in wide momentum space. This hybridization disappears when inter-layer hoppings are neglected: Then, both xy (FS1) and xy (FS2) show cusps at $\theta = \pi/4$, and xz (FS2) suddenly drops to almost zero for $\theta \geq \pi/4$. In (f), we explain the origin of nodal gap based on the fully-gapped d -wave solution in the single-Fe model^{21–23}: By introducing inter-layer hoppings, two elliptical e-pockets with positive and negative Δ in the unfolded BZ are hybridized to form FS1 and 2 with four-fold symmetry. As a result, nodal lines inevitably emerge on FS1 and 2, at least near the $|k_z| = \pi$ plane.

Here, we study the ten-orbital HH model using the RPA. As for the Coulomb interaction, we consider the intra-orbital term U , the inter-orbital term U' , Hund's coupling or pair hopping J , and assume the relation $U = U' + 2J$ and $J = U/6$. In addition, we consider the e -ph interaction due to Fe-ion optical phonons; the phonon-mediated e - e interaction ($-g$) and its matrix elements are presented in Ref. 4. Hereafter, we perform the RPA on the two-dimensional planes for $k_z = 0, \pi/2$, and π .

For $n = 6.5$ and $k_z = 0$, the critical value of g for the orbital-density-wave (ODW) is $g_c = 0.23$ eV for $U = 0$, and the critical value of U for the spin-density-wave (SDW) is $U = 1.18$ eV for $g = 0$. These values change only $\sim 2\%$ for different k_z . The obtained U - g phase diagram is very similar to Fig. 2 in Ref. 4, irrespective of the absence of h-pockets in KFe_2Se_2 . The reason would be (i) the density-of-states (DOS) in KFe_2Se_2 is about 1eV^{-1} per Fe, which is comparable with other iron pnictides, and (ii) the nesting between e-pockets is rather strong because of their square-like shape. Figure 2 (a) shows the total spin susceptibility $\chi^s(\mathbf{q}, 0)$ at $U = 1.1$ eV and $g = 0$ for $k_z = 0$ plane. χ^s is given by the intra-orbital nesting, and its peak position is $\mathbf{q} \approx (\pi, 0.4\pi)$, consistently with previous studies^{21–23}. The obtained incommensurate spin correlation is the origin of the d -wave SC gap. Figure 2 (b) shows the off-diagonal orbital susceptibility $\chi_{yz,xy;yz,xy}^c(\mathbf{q}, 0)$ for the $k_z = 0$ plane at $U = 0$ and $g = 0.22$ eV; its definition is given in Refs. 3–5. It is derived from the inter-orbital nesting between xz and xy , and its peak position is $\mathbf{q} \approx (0.7\pi, 0.4\pi)$. Note that the peak position of $\chi_{xz,xy;xz,xy}^c$ is $\mathbf{q} \approx (0.4\pi, 0.7\pi)$. The obtained strong spin- and orbital-correlations are the origin of the d -wave and s_{++} -wave SC states.

In the following, we solve the linearized gap equation to obtain the gap function, by applying the Lanczos algorithm to achieve reliable results. In the actual calculation results shown below, we take 64×64 \mathbf{k} -point meshes and 512 Matsubara frequencies. First, we study the spin-fluctuation-mediated SC state for $U \lesssim U_c$ by putting $g = 0$. Figures 3 (a)-(c) show the gap functions of the d -wave solution at $T = 0.03$ eV for $k_z = 0, \pi/2$,

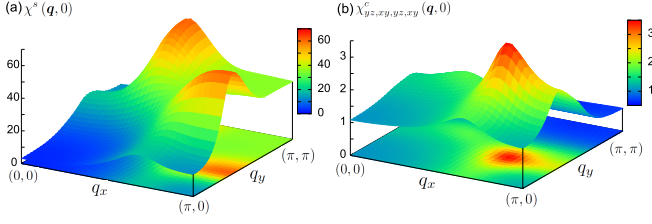


FIG. 2: (color online) (a) $\chi^s(\mathbf{q}, 0)$ for $U = 1.1$ eV and $g = 0$, and (b) $\chi^c_{yz,xy;yz,xy}(\mathbf{q}, 0)$ for $U = 0$ and $g = 0.22$ eV.

and π , respectively. In case of $U = 1.1$ eV, the eigenvalue λ_E is 0.61 for (a), 0.63 for (b), and 0.62 for (c); the relation $\lambda_E \geq 1$ corresponds to the SC state. They are relatively small since the SC condensation energy becomes small when the SC gap has complicated nodal line structure. On the (c) $k_z = \pi$ plane, the nodal lines are along $\theta = \pi/4$ and $3\pi/4$ directions, consistently with the basis of B_{1g} representation in Fig. 1 (d). These nodes move to near the BZ boundary, $\theta = 0$ and π , on the (b) $k_z = \pi/2$ plane, and they deviate from the FSs on the (a) $k_z = 0$ plane. As results, the nodal gap appears for $\pi/2 < |k_z| < 3\pi/2$ in the whole BZ $|k_z| \leq 2\pi$.

We also obtain the s_{\pm} -wave state, with the sign reversal of the SC gap between e-pockets and the “hidden h-pockets below the Fermi level” given by the valence bands 5,6. The obtained solution is shown in Fig. 3 (d) for $k_z = \pi$. Interestingly, the obtained eigenvalue is $\lambda_E = 0.99$ for $U = 1.1$ eV, which is larger than λ_E for d -wave state in Fig. 3 (a)-(c). Such large λ_E originates from the scattering of Cooper pairs between e-pockets and the “hidden h-pockets”, which was discussed as the “valence-band Suhl-Kondo (VBSK) effect” in the study of Na_xCoO_2 in Ref. 26.

Here, we analyze the T -dependence of λ_E based on a simple two-band model with inter-band repulsion: The set of gap equations is given by²⁶ $\lambda_E \Delta_h = -V N_e L_e \Delta_e$ and $\lambda_E \Delta_e = -V N_h L_h \Delta_h$, where $V > 0$ is the repulsive interaction between e- and h-pockets, and $N_{e,h}$ is the DOS near the Fermi level. When (i) the top of the h-pocket is well above the Fermi level, $L_e = L_h = \ln(1.13\omega_c/T)$, where ω_c is the cutoff energy. Thus, the eigenvalue is given as $\lambda_E = V\sqrt{N_e N_h} \ln(1.13\omega_c/T) \propto -\ln T$, similar to single-band BCS superconductors. On the other hand, when (ii) h-pocket is slightly below the Fermi level, $L_h = (1/2) \ln(\omega_c/|E_h|)$, where $E_h < 0$ is the energy of the top of h-band²⁶. Thus, the eigenvalue is given as $\lambda_E = V\sqrt{N_e N_h L_h} \sqrt{\ln(1.13\omega_c/T)} \propto \sqrt{-\ln T}$. Therefore, in case (ii), the T -dependence of λ_E is much moderate. In fact, as shown in Fig. 4 (a), λ_E for d -wave state increases monotonically with decreasing T , while λ_E for s_{\pm} -wave state saturates at low temperatures. This result suggests that the d -wave state overcomes the s_{\pm} -wave state at $T_c \sim 30\text{K}$ in $\text{K}_x\text{Fe}_2\text{Se}_2$. Although T_c in the s_{\pm} -wave state is ~ 0.06 eV in Fig. 4 (a), it is greatly reduced by the self-energy correction that is absent in the RPA⁵.

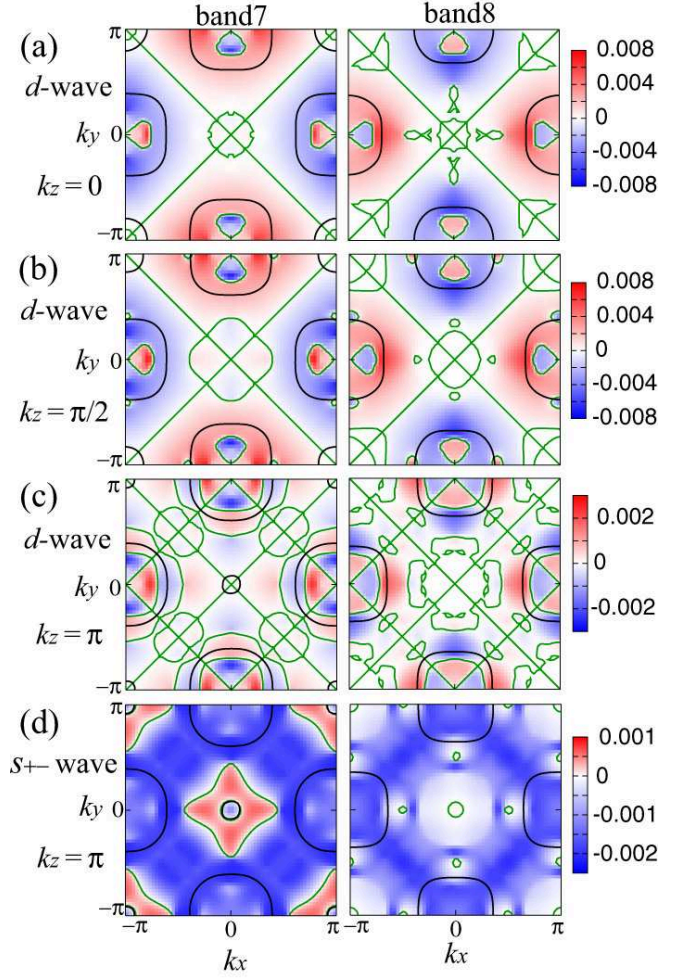


FIG. 3: (color online) SC gap functions for outer FS (FS2) on band 7 and inner FS (FS1) on band 8: d -wave gap functions on the (a) $k_z = 0$, (b) $k_z = \pi/2$, and (c) $k_z = \pi$ planes. Black and green (gray) lines represent the FSs and gap nodes. (d) s_{\pm} -wave gap function on the $k_z = \pi$ plane.

We discuss the VBSK effect for s_{\pm} wave state in more detail: According to inelastic neutron scattering measurement of $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$ ¹⁰, the characteristic spin-fluctuation energy is $\omega_{\text{sf}} \sim 100\text{K}$ just above $T_c \sim 30\text{K}$. If we assume a similar ω_{sf} in KFe_2Se_2 since T_c is close, we obtain the relation $\omega_c \sim \omega_{\text{sf}} \ll |E_h|$ in KFe_2Se_2 . Since L_h is a monotonic decrease function of $|E_h|/\omega_c$ and $L_h < 1$ for $-E_h/\omega_c > 0.15$, we consider that d -wave state overcomes the s_{\pm} -wave state in KFe_2Se_2 , as far as the spin-fluctuation mediated superconductivity is considered. Although high- T_c s_{\pm} -wave state might be realized for $|E_h|/\omega_c < 0.1$, then the realized T_c will be very sensitive to E_h or the filling n ²⁶.

Now, we study the s_{++} -wave state due to orbital fluctuations on the $k_z = 0$ plane with $n = 6.5$. In Fig. 4 (b), we show the α_c -dependence of λ_E at $T = 0.03$ for the s_{++} -wave state with $U = 0$, and the α_s -dependence of λ_E for the d -wave state with $g = 0$. Here, α_c (α_s)

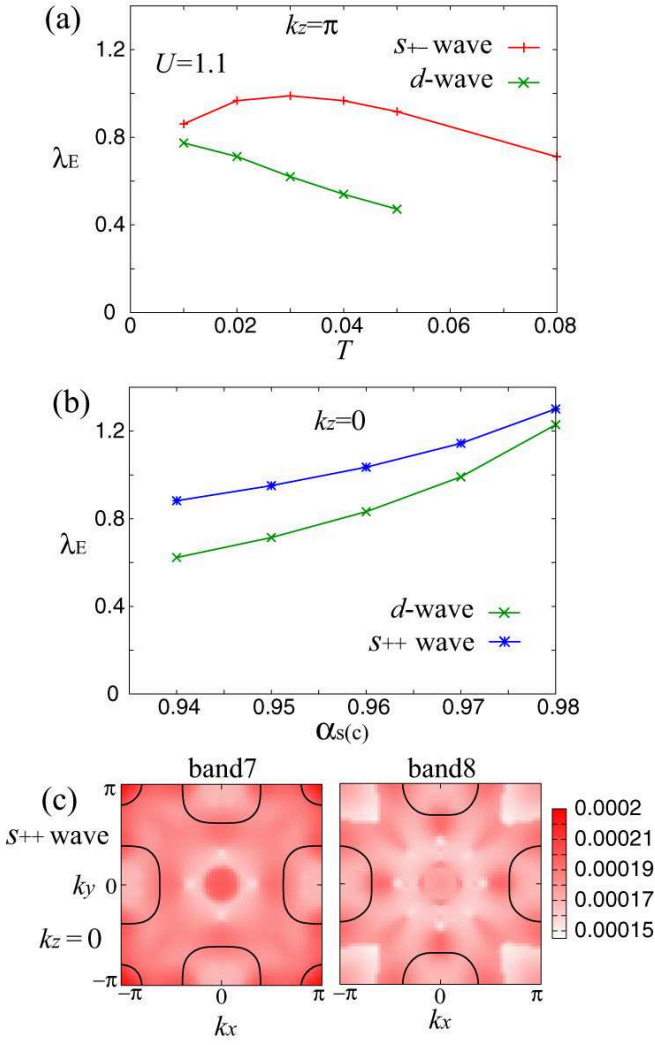


FIG. 4: (color online) (a) T -dependence of λ_E for d - and s_{\pm} -wave states. λ_E at $T = 0.01$ eV is underestimated because of the shortage of \mathbf{k} - and Matsubara-meshes. (b) α_s - (α_c -) dependence of λ_E for d -wave (s_{++} -wave) state at $T = 0.03$ eV. (c) SC gap functions for s_{++} -wave state.

is the charge (spin) Stoner factor introduced in Ref. 3; $\alpha_c = 1$ ($\alpha_s = 1$) corresponds to the ODW (SDW) state. In calculating the s_{++} -wave state, we use rather larger

phonon energy; $\omega_D = 0.15$ eV, considering that the calculating temperature is about ten times larger than the real T_c . The SC gap functions for s_{++} -wave state are rather isotropic, as shown in Fig. 4 (c). However, the obtained SC gap becomes more anisotropic in case of $U > 0^5$.

We stress that the RPA is insufficient for quantitative study of λ_E since the self-energy correction Σ is dropped: In Ref. 5, we have studied the present model based on their FLEX approximation, and found that the critical region with $\alpha_c \gtrsim 0.95$ is enlarged by the inelastic scattering $\gamma = \text{Im}\Sigma$. Also, the γ -induced suppression in λ_E for d - or s_{\pm} -wave states is more prominent than that for s_{++} -wave state, since γ due to spin fluctuations is larger than that due to orbital fluctuations⁵.

Recently, we found the paper by Mazin²⁷, in which the k_z dependence of the nodal d -wave gap in Fig. 4 corresponds to Figs. 3 (a)-(c) in the present work.

In summary, we studied the mechanism of superconductivity in KFe_2Se_2 based on the ten-orbital HH model without h-pockets. Similar to iron-pnictide superconductors, orbital-fluctuation-mediated s_{++} -wave state is realized by small dimensionless e -ph coupling constant $\lambda = gN(0) \sim 0.2$. We also studied the spin-fluctuation-mediated d -wave state, and confirmed that nodal lines appear on the large e -pockets, due to the hybridization between two e -pockets that is inherent in 122 systems. Therefore, careful measurements on the SC gap anisotropy is useful to distinguish these different pairing mechanisms. Study of impurity effect on T_c is also useful since d -wave (and s_{\pm} -wave) state is fragile against impurities.

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